SELF-PROTECTION ANALYSIS OF DENATURED THORIUM-PLUTONIUM FUEL

By

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ABSTRACT

With growing demands for commercial nuclear power, there is also a growing need for better energy efficiency from nuclear power reactors. In order to reach a high burnup up to 100 MWd/kg, previous research has examined the use of thorium-plutonium mixed-oxide fuel as a potential candidate for this high-burnup goal. Though the neutronics studies have looked upon this fuel type favorably, the purpose of this paper is to investigate the self-protection capabilities of this fuel type, for anti-proliferation purposes. In particular, there were two proliferation-resistance methods that were analyzed. First, this study examined the time-dependant dose-rate of the spent fuel caused by the decay of the isotope uaranium-232, which releases a high-energy gamma of 2.6 MeV. Next, this study examined the possibility of denaturing the fuel with depleted uranium in order to dilute the weapons-usable isotope uranium-233 in the spent fuel. The U-232 dose rate was also calculated for the denatured case. Ultimately, the study found that there was a negligible different in the amount of time that it takes for either fuel type to become self-protective. The denatured case showed that it requires much more plutonium than the undenatured case in order to ensure that there is not sufficient weapons-usable U-233 in the discharged fuel.

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1. Introduction

On July 1, 1968, a collection of nations came together to sign the Non-Proliferation Treaty, otherwise known as NPT. The treaty came in response to a division between those nations who were building nuclear weapons, and those who were not. The Nuclear Weapons States were actually more supportive of peaceful nuclear technology and actively executed more research for this purpose. Therefore, the treaty was created in order to allow the rest of the world to benefit from peaceful nuclear technology while the Nuclear Weapons States could protect their right to weapons possession. Furthermore, the treaty allowed the Non-Nuclear Weapons States to have access to nuclear power so long as they promised to not develop their own weapons. Lastly, the treaty led to the creation of the International Atomic Energy Agency (IAEA) to regulate this agreement made by the collection of nations participating.

However, the International Atomic Energy Agency has warned of the black market that exists for gas centrifuges and nuclear fuel. This black market is thought to exist in many countries including Pakistan, Malaysia, Iran, and North Korea—all countries known to have possession of nuclear explosives. Therefore, the possibility of enriched uranium falling into the wrong hands is definitely a possibility, so nuclear power regulations must be imposed to account for that threat. One such regulation is the limitation of uranium enrichment to very low levels, with 20% being the maximum weight percentage allowed for any reactor type. The present de facto limit for all power reactors is 5%. However, with growing demands for nuclear energy also comes a growing need for better energy efficiency from commercial power reactors. In other

words, there is a need for higher enrichment of fissile material (for higher burnup) in order to extract more energy from the fuel, and achieve longer fuel cycles as well as less frequent refueling.

Some light water reactors (LWRs) today use a uranium-plutonium mixed-oxide (MOX) fuel for power generation. This mixed fuel was used to replace the conventional uranium-dioxide (UO_2) because of the large amounts of plutonium available from spent nuclear fuel. Previous research has demonstrated, however, that if this uranium-plutonium mixed-oxide fuel were enriched with fissile material to higher levels then it would create a positive void coefficient in the reactor. Currently, the limit for enrichment of plutonium in MOX fuel is about 12% for this reason. A positive void coefficient is undesirable for operation purposes because it means that that the reactivity increases as the void content inside the reactor increases due to increased boiling or loss of coolant. Therefore, a better option was introduced by replacing the uranium with thorium in the mixed-oxide fuel in order to eliminate this positive void coefficient concern in reaching higher burnup levels.

Furthermore, the replacement of uranium with thorium is desirable for other reasons. First, it addressed the concern that uranium reserves would start to diminish in the coming years. Second, the use of thorium proved to be more efficient at depleting plutonium in the MOX fuel. This was important because plutonium was considered the most significant threat for nuclear proliferation, so it was necessary to find ways to decrease the overall mass of plutonium and not produce any significant amounts of additional plutonium (which the uranium in the MOX fuel actually was doing). For example, a study showed that the uranium-plutonium MOX can deplete about 544 kg of

plutonium per year, while the thorium-plutonium can deplete about 800 kg of plutonium per year [1]. Lastly, the thorium-plutonium MOX fuel produced a transmutation chain product, uranium-232, which emits a very high-energy gamma ray. This meant that the fuel would be self-protecting by making it more vulnerable to radiation detection methods as well as more difficult to handle (from a radiation safety standpoint).

2. Statement of Problem

2.1 Background Information

Present day discharge burnups for UOX fuel are about 50 MWD/kg. In trying to reach high burnup up to 100 MW-day/kg, there are three major obstacles: the positive-void coefficient when using a uranium-plutonium MOX fuel, temperature restraints in the fuel bundle cladding material, and most importantly the proliferation concern of producing weapons-usable fissile material. As stated above, the issue with the positive-void coefficient has already been addressed by the replacement of uranium with thorium in the MOX fuel. A recent study analyzed various reactivity coefficients such as moderator temperature coefficient, Doppler coefficient, void coefficient, and soluble boron worth for MOX fuel and for thorium-plutonium fuel. The conclusion was that the thorium-plutonium fuel was more advantageous because it had a negative Doppler coefficient and moderator temperature coefficient [2].

The next obstacle involving the cladding material is due to the current material used in commercial nuclear power plants, zirconium alloy. A previous study has investigated the use of thorium-plutonium fuel, but only up to a burnup level of 60 MWd/kg. This was because the Nuclear Regulatory Commission has set a regulation limiting LWR burnup at 62 MWd/kg due to this cladding issue. After withstanding a burnup of 62 MWd/kg, the ductility of the zirconium alloy cladding is reduced by a factor of five [3]. This happens because as temperature of the reactor increases, the fuel pellet and cladding both experience thermal expansion while there is also a continual buildup of fission product gases increasing the pressure between the fuel and the cladding. This

buildup of pressure imposes heavy strain on the clad material and also leads to the buildup of an oxide layer making the zirconium alloy more brittle. Finally, the diffusion of hydrogen into this gap creates zirconium hydride (ZrH_2), which makes the cladding even more brittle and more likely to crack under transient situations [3].

Current research, however, is trying to address this concern by introducing a new type of cladding material called Silicon Carbide (SiC). The idea is that this cladding might be more suitable for reaching higher burnup because it is a ceramic material. The fuel is not allowed to come into contact with the cladding because it is brittle material, so a wider gap is needed. Therefore, the larger gap thickness will allow the fuel to reach higher temperatures. Additionally, the thermal conductivity of silicon carbide is lower than zirconium alloy; thus, the buildup of fission gases will be able to diffuse more efficiently [4]. Previous research has shown that silicon carbide has the potential of being able to withstand burnup levels up to 100 MWd/kg without damage to the cladding or the fuel.

Finally, the last concern in reaching high burnup levels is regarding the issue of proliferation threat in producing fissile material. Typically, weapons are made using highly-enriched uranium (HEU) or plutonium-239 as the primary fuel. Highly-enriched-uranium must be enriched above 20% in order to be weapons-usable, which is partly the reason why it is currently the upper enrichment limit for research reactors. Plutonium-239 does not exist in nature because it is only produced in reactors by neutron absorption in U-238 (with subsequent beta decay). In the proposed thorium-plutonium MOX fuel cycle, however, there is a third fissile isotope that can pose a significant risk: uranium-233. It is produced from the neutron absorption of thorium-232, which is the main fertile

isotope in this fuel type. These three isotopes—U-235, Pu-239, and U-233—are all potential proliferation risks because they all have large thermal-neutron fission cross-sections. Therefore, if any of these three isotopes were to fall into the wrong hands, it could lead to serious illegal weapons fabrication. The purpose of this report is to examine two particular methods of making this thorium-plutonium fuel self-protective against attempts at illegal activity for weapons purposes.

2.2 Uranium-232 and Dilution of Uranium-233

The first proposed form of self-protection involves the presence of the isotope uranium-232 in the spent fuel—a product of a high-energy (n, 2n) reaction in uranium-233. This uranium isotope is significant because it releases a very powerful gamma ray of energy 2.6 MeV from the decay of thallium-208, a daughter product in the decay of U-232 [5]. Figure 1 illustrates the decay chain of U-232 along with the half-life of all the subsequent radioisotopes, ending with the stable isotope lead-208.

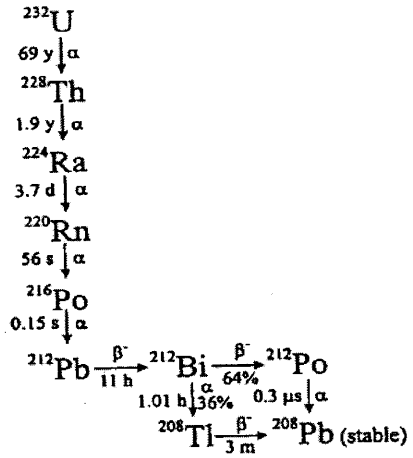

Figure 1. Decay Chain of Uranium-232

This powerful gamma emission is very important for two reasons. First, this makes the fuel very easy to detect with radiation security equipment. Second, this kind of ionizing radiation can make it very difficult to handle such material. In trying to build a nuclear weapon, the process requires the chemical separation of the fissile material U-233 from the rest of the MOX fuel. But with the U-232 isotope present, it would be very difficult to extract the material due to the high-energy penetrating gamma rays emitted from the fissile material.

The next proposed form of self-protection involves the denaturing of the thorium-plutonium fuel using depleted uranium in order to dilute the weapons-usable isotope U-233. According to one particular study, the discharged fuel is considered non-weapons usable so long as the proliferation index is less than 0.12 [6]. This is defined as:

$$\frac{\text{Weight of } {}^{233}\text{U} + 0.6 \times \text{Weight of } {}^{235}\text{U}}{\text{Total Weight of Uranium}} < 0.12 \qquad (1)$$

By denaturing the thorium-plutonium fuel with a small admixture of depleted uranium (0.25% enriched), the amount of weapons-usable U-233 will be depleted to low amounts therefore rendering the discharged fuel resistant to proliferation attempts. For the purposes of this study, depleted uranium was chosen over natural uranium because of availability and also because of cost.

2.3 Goals

In order to present deliverables with concrete analysis, the following is a list of the main tasks investigated in this study:

1. **Uranium-232 dose rate in un-denatured fuel**— In order to quantify the degree to which uranium-232 is making the undenatured fuel self-protective, the maximum dose rate will be calculated in units of rem/hour. This number will be important in understanding the radiological properties of highly enriched thorium-plutonium MOX fuel used to reach a discharge burnup of 100 MWd/kg.
2. **Thorium, Plutonium, Uranium concentrations**— In determining the initial concentrations of each individual heavy metal, there are two boundary conditions that must be met: a discharge burnup of 100MWd/kg, and a proliferation index approximately under 0.12.
3. **Uranium-232 dose rate in denatured fuel**— Though already self-protected in theory, the dose rate from U-232 in the denatured fuel will also be calculated. This will be compared to the dose rate of the un-denatured case in order to see the effect that the uranium admixture has on the production of U-232 in the discharged fuel.

2.4 Benchmarks

Before making any calculations, there are a couple of benchmarks that must be established in order to make accurate comparisons further in the study. First, before analyzing the initial composition of the heavy metals in the thorium-plutonium-uranium MOX fuel for reaching higher burnup levels, there needs to be a benchmark for the initial composition of uranium dioxide in conventional power operation. In other words, the U-235 enrichment will reach higher levels than usual for burnup levels up to 100 MWd/kg. Figure 2 shows how the enrichment of uranium dioxide follows the linear reactivity model for discharge burnup.

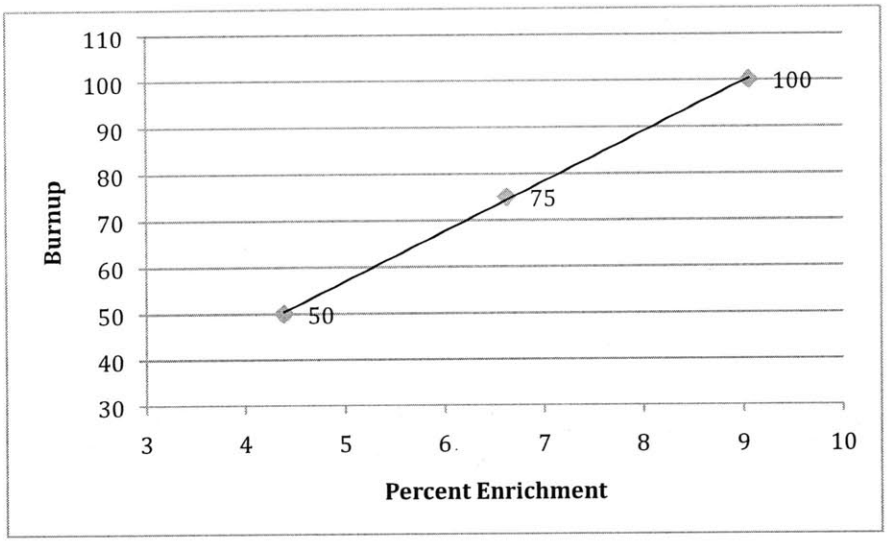

Figure 2. The Linear Reactivity Model of Uranium Dioxide Fuel.

Assuming a leakage reactivity of 3%, the U-235 enrichment for discharge burnups of 50MWd/kg, 75 MWd/kg, and 100 MWd/kg were calculated to be 4.39%, 6.625%, and 9.06%, respectively.

Next, a benchmark is needed for the initial concentration of the heavy metals in undenatured thorium-plutonium MOX fuel when reaching high discharge burnup. Figure 3 shows the linear reactivity behavior of the fuel for a single-batch burnup of up to 75 MWd/kg. The figure is originally from a study comparing conventional MOX fuel to thorium-plutonium MOX fuel [7]. By extrapolation, the plutonium enrichment for discharge burnup levels of 50 MWd/kg, 75 MWd/kg, and 100 MWd/kg are approximately 9%, 13%, and 17%.

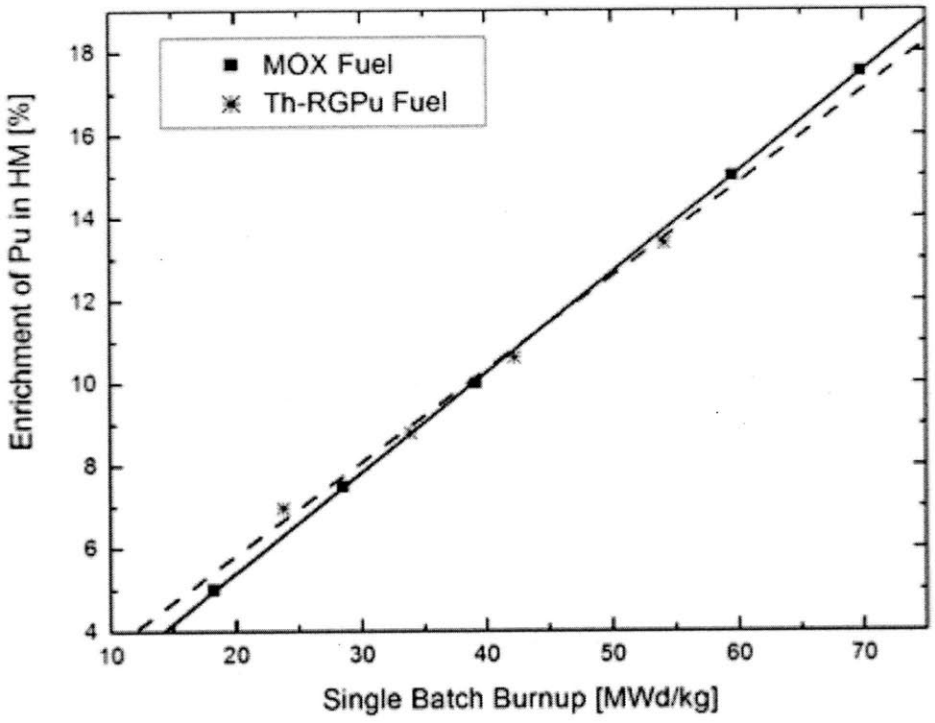

Figure 3. Linear Reactivity Model for MOX and Th-RGPu [7].

3. Procedures

3.1 Calculating Dose Rate of Discharged Undenatured Fuel

The first step in calculating the dose rate was determining the composition of the fuel at the discharge burnup of 100 MWd/kg. This was done using the simulation code CASMO—a two-dimensional, multi-group transport code that is used for simulations of BWR/PWR assemblies or pin cells. It relies on 70 energy groups to calculate all the neutronics parameters in the simulations, using an internal library called JEFF-2.2 [8]. For the purposes of this study, only one pin cell was used for the simulations. The geometry and operating conditions of a typical PWR pin cell are listed in Table 1.

Table 1. Typical Operation Conditions of PWR Pin Cell

Fuel Temperature [K]	900
Coolant Temperature [K]	583.1
Operating Pressure [MPa]	15.5
Core Power Density [kW/L]	104.5
Fuel Pellet Diameter [mm]	8.192
Gap Thickness [mm]	0.082
Fuel Cladding Outer Diameter [mm]	9.500
Fuel Element Pitch [mm]	12.6

A visual image of the pin cell is shown in Figure 4.

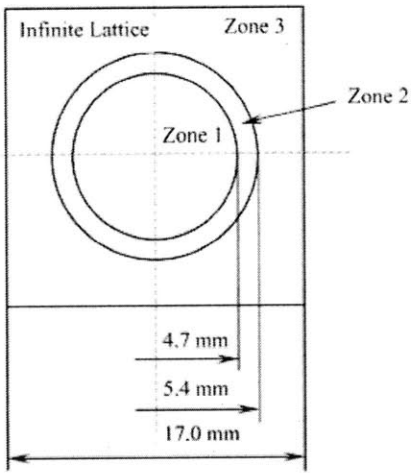

Figure 4. Geometric schematic of PWR pin cell.

As stated in section 2.4, the plutonium enrichment for the undenatured case achieving a discharge burnup of 100 MWd/kg was determined to be 16.3%, with the rest of the initial heavy metal composed of thorium-oxide (83.7%). In running CASMO for this case, the theoretical densities of plutonium oxide (PuO_2) and thorium oxide (ThO_2) were assumed to be 11 g/cm^3 and 9.6 g/cm^3, respectively, with a total averaged theoretical density of 9.8 g/cm^3 [9]. However, only 96% of this theoretical density was used as the input density for the simulation. For the composition of the ThO_2, only the Th-232 isotope was used. For PuO_2, however, the composition is listed in Table 2.

Table 2. Isotopic Composition of Plutonium

Nuclide	Weight Percent
Pu-238	2%
Pu-239	53%
Pu-240	25%
Pu-241	15%
Pu-242	5%

After running the simulation to a burnup of 100 MWd/kg, the final isotopic composition of the fuel was extracted. In particular, the weight percentage of U-233 in the discharged fuel was recorded for further calculation. An important assumption used was that the total weight of one fuel assembly was 450 kg of heavy metal mass. Therefore, the total weight of U-233 at the discharge burnup was calculated by multiplying the weight percentage of U-233 by the total heavy metal mass of 450 kg, and then reduced by a factor of two to account for discrepancies in U-232 distribution in a fuel rod. According to a recent study, the ratio of U-232 production to U-233 production in a typical PWR is approximately 7.267×10^{-3} [10]. From this ratio, the total initial weight of the isotope U-232 was deduced.

Next, figure 5 shows the total dose rate for 1 gram of U-232 as a function of time. The dose rate continues to grow until it reaches a peak at around 9 years of decay, with a maximum dose rate of about 21 rem/hour per gram of U-232 initial.

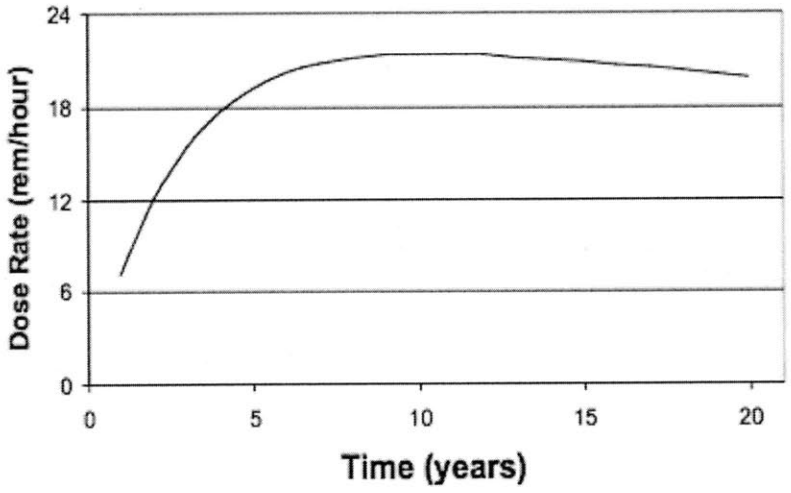

Figure 5. Dose Rate for 1 gram of U-232 initial [10].

To calculate the peak dose rate for discharged undenatured thorium-plutonium MOX fuel, the total initial weight of U-232 was multiplied by this peak value of 21 rem/hour per gram of U-232 initial.

3.2 Calculating the Concentrations of Plutonium, Uranium, Thorium

As stated in section 2.3, there were two main boundary conditions in calculating the concentrations of the heavy metals used in the denatured case: a discharge burnup of 100 MWd/kg, and a Proliferation Index approximately equal to but less than 0.12. In running CASMO for this case, the theoretical densities of PuO_2, ThO_2, and UO_2 were assumed to be 11 g/cm^3, 9.6 g/cm^3, and 10.5 g/ cm^3, respectively, with a total averaged theoretical density of 9.98 g/cm^3. Again, only 96% of this theoretical density was used as the actual density in the input. The discharge burnup is defined as,

$$BU_n = \frac{2n}{n+1} \cdot BU_1. \tag{2}$$

Assuming a fuel cycle with n=3 batches, the discharge burnup becomes 1.5 * BU_1.

Through trial and error in adjusting the amount of plutonium and depleted uranium used in the CASMO simulation, the aim was to find a combination that achieved a single-batch burnup close to 66.67 MWd/kg, implying a discharge burnup of 100 MWd/kg according to equation 3. The single-batch burnup is defined as the equilibrium burnup where the reactor remains critical. In other words, it is the burnup at which k_{inf} is at a value of 1.03 (the 0.03 accounts for the leakage reactivity worth). After finding a

right match, the fuel composition was extracted from the discharged fuel in order to verify the proliferation index using equation 1.

3.3 Calculating Dose Rate of Discharged Denatured Fuel

This step was very similar to the process used to calculate the dose rate for the discharged undenatured fuel described in section 3.1. Using the fuel composition found in section 3.2, the weight percentage of U-233 was recorded and used to calculate the total initial weight of U-232 (using the same assumption that the total heavy metal mass was originally 450 kg). To calculate the peak dose rate for discharged denatured thorium-plutonium MOX fuel, the total initial weight of U-232 was multiplied by the peak value of 21 rem/hour per gram of U-232 initial. Next, the total time to become self-protective was calculated for both fuel types. This was done by examining the amount of time (based on the correlations described above) that it took to reach a dose rate of 100 rem/hour, which is the minimum dose-rate needed for self-protection.

4. Results

4.1 Uranium-232 dose rate in undenatured fuel

At the end of its life cycle, the undenatured fuel contains a total weight percent of 1.973% of the fissile isotope U-233. Assuming a total heavy metal mass of 450 kg, this results in a total mass of 8.87 kg of U-233. After multiplying this number by the U-232/U-233 ratio and reducing by a factor of two, the resulting weight of uranium-232 in the discharged fuel is approximately 32.26 grams. Using the correlation illustrated in figure 5, the peak dose rate from U-232 in spent thorium-plutonium MOX fuel is approximately **677.46 rem/hour**, for a radius of 1 meter after a time span of 9 years. This is well above the 100 rem/hour self-protection standard needed for resistance to proliferation.

4.2 Thorium, Plutonium, Depleted Uranium Concentrations

Figure 6 shows how the value of k_{inf} changes with increasing burnup for both the denatured and undenatured case. For both cases, the value of the neutron multiplication factor is equal to 1.03 at the point where the single-batch burnup is equal to 66 MWd/kg. Interestingly enough, the curves intersect at the point where k_{inf}=1.03, with the denatured case having a lower slope than the denatured case. This is probably due to the fact that the admixture of uranium makes it more efficient at breeding fissile material, since the U-238 in the material keeps interacting with neutrons and producing additional Pu-239 in the fuel.

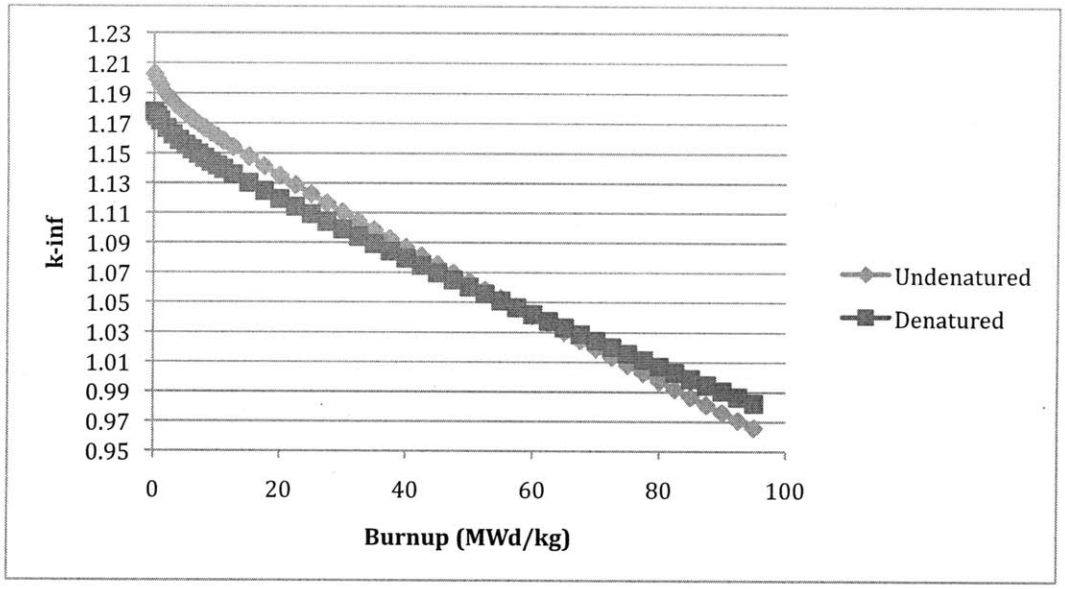

Figure 6. Undenatured versus Denatured.

Table 3 shows the composition of both fuel types in addition to the proliferation index of the denatured case. The proliferation index does not apply to the undenatured case because it is only used for cases where U-233 is isotopically diluted with U-238. In order to maintain the proliferation index for the denatured case under 0.12, it requires a significant amount of more plutonium than the undenatured case.

Table 3. Isotopic Composition of Both Fuel Types

	Plutonium	**Thorium**	**Uranium**	**P.I.**
Denatured	19.81%	15.90%	64.29%	0.11
Undenatured	16.80%	83.20%	0	N/A

4.3 Uranium-232 Dose Rate in Denatured Case

At the end of its life cycle, the denatured fuel contains a total weight percentage of 1.741% of U-233, less than the amount in the undenatured case. Assuming a total heavy metal mass of 450 kg, this results in a total mass of 7.83 kg of U-233. After multiplying this number by the U-232/U-233 ratio and reducing by a factor of two, the resulting weight of uranium-232 in the discharged fuel is approximately 28.47 grams. Using the correlation illustrated in figure 5, the peak dose rate from U-232 in spent denatured fuel is approximately **597.8 rem/hour**, for a radius of 1 meter after a time span of 9 years. This dose rate is also well above the 100 rem/hour self-protection standard needed for resistance to proliferation. Therefore, both the denatured case and undenatured case demonstrate a fair ability to resist proliferation attempts simply by the presence of the high-energy gamma from uranium-232.

Using the same correlation from figure 5, table 4 shows the amount of time that it takes for both cases to become self-protective assuming the initial U-232 amount described above. A fuel bundle is considered self-protective if the total measured dose rate is above 100 rem/hour per gram of U-232 initial.

Table 4. Beginning of Self-Protection

	U-232 Initial	**Beginning of Self-Protection**
Undenatured	32.26 grams	21 weeks
Denatured	28.47 grams	26 weeks

As expected, the addition of depleted uranium causes the dilution of weapons-usable U-233, therefore reducing the amount of U-232 present in the spent fuel. This, in turn, means that it takes longer for the denatured fuel type to become self-protective. However, it is only a matter of 5 weeks difference.

5. Conclusions

5.1 Discussion

The study presented in this report demonstrated that thorium-plutonium mixed-oxide fuel denatured with depleted uranium could very well be a viable alternative to conventional power reactor fuel limited to 5% enrichment. In trying to reach a high burnup of 100 MWd/kg, there are three main obstacles for the typical uranium-plutonium mixed oxide fuel. First, there is the problem caused by positive void coefficient in the fuel. This was easily solved by replacing the uranium with thorium as the main fertile fuel. Second, the cladding made of zirconium alloy can only withstand burnup up to 62 MWd/kg. Therefore, an initiative was created with the aim to find a cladding replacement that can withstand higher temperatures. A number of studies at MIT have focused on silicon carbide as a possible candidate for this new cladding. Finally, there is the issue regarding proliferation concern and making the spent fuel resistant to proliferation threats.

This study has shown that the undenatured case and the denatured case both demonstrate a substantial ability to be self-protective by the presence of strong gamma emissions from the decay of U-232. Because the depleted uranium helps dilute the amount of U-233 in the denatured case, the peak dose rate is lower than that of the undenatured case. Therefore, it takes about five more weeks for the denatured fuel to become self-protective. In the larger scheme of illegal activity, however, this is a relatively small difference in time given that it takes months to transport the spent fuel and proceed to chemically separate the isotopes. Therefore, both the undenatured and

denatured cases have demonstrated the ability to resist proliferation attempts solely based on the high dose-rate and easy detectability.

5.2 Suggestions for Future Research

When it comes to choosing between an undenatured fuel type and a denatured fuel type, the question really comes down to the economics. Table 3 shows that the denatured fuel requires a much heavier concentration of plutonium in the fuel than the undenatured fuel (for a discharge burnup of 100 MWd/kg). A future project could investigate the economic implications of requiring more plutonium while also using uranium to dilute the weapons-usable isotope U-233. Ultimately, both cases are self-protective so there might be no need to dilute the fissile material. However, given that there is still a time window when then the fuel is not self-protective (approximately 21 weeks), then it might be more logical to use the denatured fuel type in order to ensure absolute security of the fuel. At the same time, this hypothetical study could also examine the trade-offs in using depleted uranium versus natural uranium to dilute the weapons-usable material. For example, depleted uranium might be more available and less costly; however, the use of natural uranium would allow for less use of plutonium and would aid in reducing the amount of capital costs in extracting plutonium from spent fuel.

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